

## N92-21615

MICROWAVE (EPR) MEASUREMENTS OF THE PENETRATION DEPTH  
MEASUREMENTS OF HIGH- $T_c$  SUPERCONDUCTORSN.S.DALAL\*, B.RAKVIN<sup>a)</sup>, T.A.MAHL, A.S.BHALLA<sup>b)</sup>,  
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Abstract Electron Paramagnetic Resonance (EPR) has been found to be a quick and easily accessible method for measuring the London penetration depth,  $\lambda$ , for the high- $T_c$  superconductors. The method utilizes the broadening of the EPR signal of a spin-probe compound adsorbed to the surface of the superconductor. The broadening of the EPR signal is due to the formation of the magnetic flux lattice below  $T_c$  and is measured as the second moment,  $\langle \Delta H^2 \rangle$ .  $\langle \Delta H^2 \rangle$  is fitted to the Brandt equation for a simple triangular lattice:  $\langle \Delta H^2 \rangle = 0.00371 \lambda_0^2 [1 - (T/T_c)^4]^{-1/2}$ . This method yields  $\lambda_0 = 2700 \pm 100$  Å with a  $T_c$  of 84 K for  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$  and  $2520 \pm 100$  Å with a  $T_c$  of 119 K for  $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ . The precision of  $\pm 100$  Å or better compares well with those of the more traditional techniques of  $\mu^+\text{SR}$ , neutron diffraction, and magnetic susceptibility.

INTRODUCTION

Currently several methods are in use for the measurement of  $\lambda$ , such as muon spin resonance ( $\mu^+\text{SR}$ )<sup>1,2</sup>, d.c. susceptibility<sup>3,4</sup>, neutron scattering<sup>5</sup>, and NMR<sup>6,7</sup>. However, each of these methods has some shortcomings<sup>1-6</sup>. For example, muon spin resonance measures  $\lambda$  via line broadening as a result of the flux penetration. However, one must estimate the contribution from relaxation broadening, which appears hard to do<sup>1,2</sup>. Neutron scattering needs access to a high (neutron) flux reactor and studies the bulk properties of a sample<sup>5</sup>.

Susceptibility measurements require an estimation of the lower critical field  $H_{c1}$ , since  $\lambda$  is measured via  $H_{c1} = \phi_0 / (\lambda_0^2)^{3,4,8}$ . However,  $H_{c1}$  is hard to measure for these materials<sup>9</sup>.

To our knowledge, there is only one, rather recent report<sup>10</sup> on the use of EPR spectroscopy for the measurement of  $\lambda$ . However, the measurements were only made on  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ , and over a small temperature range ( $\approx 15^\circ$ ) below  $T_c$  while measurements at lower temperatures were particularly desirable<sup>10</sup>. The present work has improved the methodology by making more detailed measurements over a wider range of temperatures, analyzing the data with a more accurate theoretical model<sup>11</sup>, and applying it to two new lattices.

#### PRINCIPLE OF THE EPR METHODOLOGY FOR MEASURING $\lambda$

The principle of the EPR methodology is similar to that proposed by Pincus et al.<sup>7</sup> in 1964 for NMR for the Type-II superconductors. The linewidth of the NMR signal is increased by the inhomogeneous magnetic fields created by the emergence of a magnetic flux lattice below  $T_c$ , when the external field  $H_0$  satisfies the condition  $H_{c1} < H_0 < H_{c2}$ . The same type of inhomogeneous broadening is expected to influence the EPR line since the flux lattice should be field independent within the magnetic field range described. Thus the EPR method is based on the measurement of the linewidths or second moment of the EPR line of a paramagnetic probe adsorbed on the surface of a Type-II superconductor. In the absence of detailed knowledge of the flux lattice in these high- $T_c$  superconductors, we shall assume a simple model of a perfect triangular lattice for a Type-II superconductor<sup>11</sup>.

Brandt<sup>11</sup> recently reported for Type-II superconductors the formula for the second moment data in the case of a perfect triangular lattice as:

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$$\langle \Delta H^2 \rangle = 0.00371 \Phi_0^2 / \lambda^4 \quad (1)$$

$$\lambda = \frac{\lambda_0}{[1 - (T/T_c)^4]^{1/2}} \quad (2)$$

where  $\Phi_0$  is the flux quantum and  $\lambda_0$  is the penetration depth at  $T = 0$ . The temperature dependence of  $\lambda$  is assumed to be described with the standard two-fluid form<sup>1,2</sup> (eq. 2).

### EXPERIMENTAL

The paramagnetic probe used for the measurements was the stable free radical Diphenylpicrylhydrazyl (DPPH). The DPPH was dissolved in acetone to  $\approx 10^{-2}$  M concentration. A few milligram sample of the superconductor was then immersed in the ( $\approx 10^{-2}$ M) DPPH solution and then dried in air. The EPR measurements were made using a Bruker ER 200D EPR spectrometer, operating in the X-band (9.5 GHz). The temperature was controlled to  $\pm 0.1$  K using an Oxford Instrument model DRC1 temperature controller. All measurements were performed with magnetic field modulation amplitudes in the range of 0.8 - 4 Oe at a frequency of 100 kHz. The microwave power was kept low ( $\sim 1$ mW) to minimize power saturation and broadening of the DPPH EPR signal. The superconductors,  $Tl_2Ba_2Ca_2Cu_3O_x$  and  $Bi_2Sr_2CaCu_2O_x$  were studied.

### RESULTS

Typical EPR spectra for the DPPH radical<sup>13,14</sup> adsorbed on both the  $Tl_2Ba_2Ca_2Cu_3O_x$  ( $T_c = 119$ K) and the  $Bi_2Sr_2CaCu_2O_x$  ( $T_c = 82$ K) superconductors are shown in Figure 1. As can be seen, the signal broadens rapidly for temperatures less than  $T_c$  for each sample. The spectra showed no significant broadening above  $T_c$ . The second

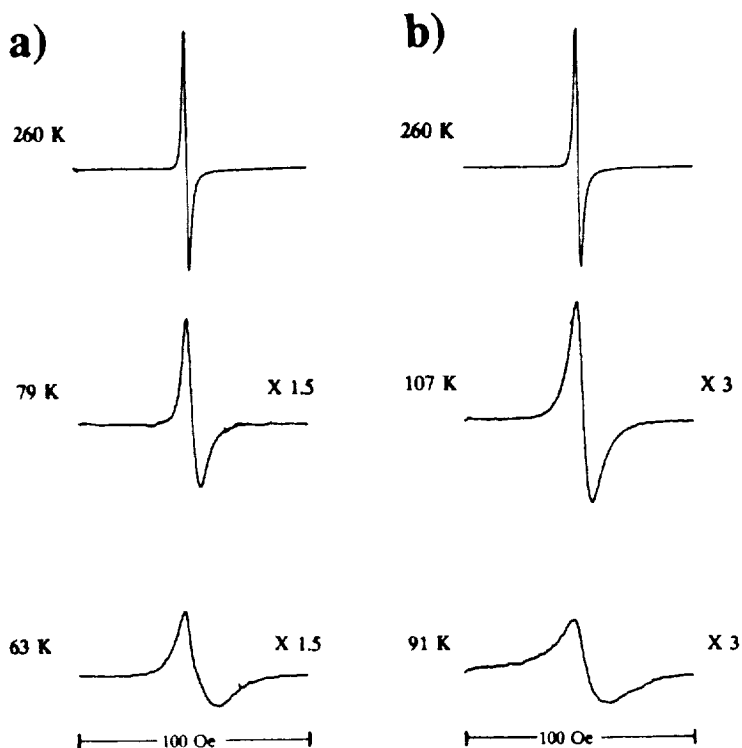


Figure 1. The EPR signals of DPPH adsorbed on (a)  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$  and (b)  $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_x$  at the indicated temperatures.

moments of the EPR signals were calculated using the equations described by Poole<sup>14</sup>. The second moment values from the experimental results are shown as symbols in Figure 2a and 2b for  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$  and  $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_x$  respectively.

The best fitting curves that were obtained are with the parameters  $\lambda_0 = 2520 \text{ \AA}$  and  $T_c = 119 \text{ K}$  for  $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_x$  and  $\lambda_0 = 2700 \text{ \AA}$  and  $T_c = 84 \text{ K}$  for  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$  as can be seen by the solid lines in Figure 2. The dashed lines in the figures indicate plots for  $\lambda_0$  differing by  $\pm 100 \text{ \AA}$  demonstrating the precision of the fitting procedure. It can be seen that all of the experimental data points lie within these lines indicating that this method can yield  $\lambda_0$  well within an precision of  $\pm 100 \text{ \AA}$ .

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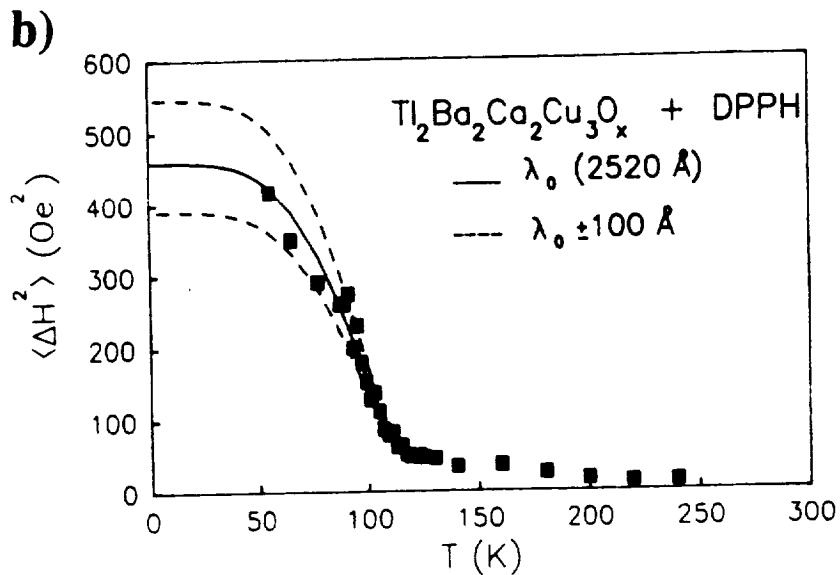
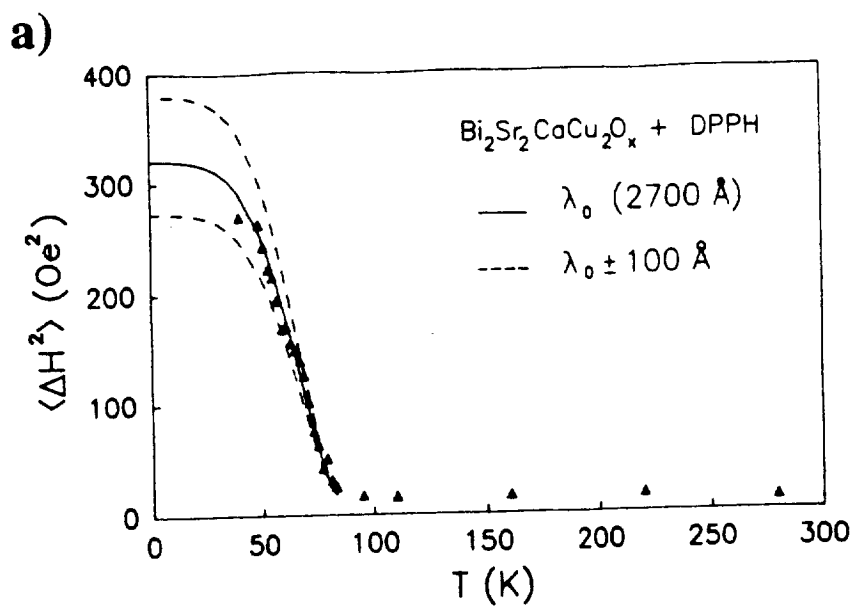


Figure 2 The second moment,  $\langle \Delta H^2 \rangle$ , vs. temperature for DPPH adsorbed on (a) Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>x</sub> and (b) Tl<sub>2</sub>Ba<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub>. The curves are fits to equations (1) and (2) in the text.

Accurate measurements of  $\lambda_0$  for these samples by the established methods of  $\mu^+$ SR and polarized neutron scattering are not available. However, the  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$  result of  $\lambda_0 = 2700 \text{ \AA}$  is in the range of values of  $1585 \text{ \AA}$  and  $3650 \text{ \AA}$  determined from critical field measurements for the related compound  $\text{Bi}_4\text{Ca}_3\text{Sr}_3\text{Cu}_4\text{O}_{16}$ <sup>8</sup>. There are to this point no literature values for the  $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ .

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